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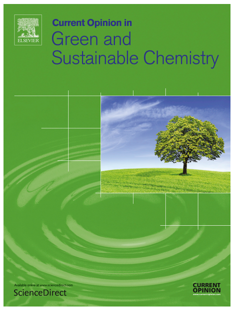

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Exploring new horizons for paper recycling: A review of biomaterials and biorefinery feedstocks derived from wastepaper

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Abstract

Paper is a perfect example of the circular economy as it remains the furthermost recycled product in Europe, creating significant environmental benefits and raw materials resources to the industry. Indeed, maintaining a consistent level of quality whilst limiting the environmental footprint of the product has become a major challenge for the industry. In this direction, paper is proving to be the promising feedstock for biorefinery and biomaterials. The future of paper recycling is slowly going beyond fibre recovery to address the needs of other industries because for the earth's environmental well-being various paper products need to be recycled and reused persistently. In this article, we outline the ambitious use of wastepaper (WP) for high-value applications such as; production of cellulose nanocrystals (CNC), composite reinforcement, high performance electrical components and biofuels.

Keywords: Wastepaper, Recycling, Cellulose, Ethanol, Hydrogen

1. Introduction

Paper recycling is a major process in the manufacturing of paper products. Europe boasts of 72% recycling rate recorded in 2012 and a 74% target is set for 2020 [1]. Although, paper can be recycled an average of 5 times, most paper products have a very short lifespan (days) which may not justify the amount of resources consumed for its production and its use in low value applications. The expectations for paper recycling rates to increase, requires the use of recycled fibers in high quality grade paper. However, these fibers become shorter and reduce the properties in swelling and flexibility which cannot meet the standards desired by customers. Furthermore, the demand for high quality paper products containing recycled fibers rely on the increasing use of chemical additives and fillers which produce large quantities of by-products posing serious environmental and economic challenges [2].

An overview of publications on paper recycling is illustrated in **Fig.1.** The authors reviewed publication overtime on Scopus database which showed that, early studies were focused on fiber recovery from wastepaper which included fiber bleaching methods and deinking technologies to allow the reuse of secondary fibers in papers, an increasing demand for deinking equipment was also noted in the early nineties. Publications on paper recycling peaked in year 2000 perhaps in response to the European declaration on paper recycling released the same year [3]. In 2001-2010 publications were still dominated by fiber recovery. Nowadays circular economy principles are becoming attractive within regulators and policy makers, as the substitution of fossil fuel based products with bio-based or biodegradable materials is strategic for mitigating climate change. Thus, the current landscape on wastepaper recycling is populated with recovery of biomaterials and bio-refinery feedstock from waste paper which will be discussed in this review.

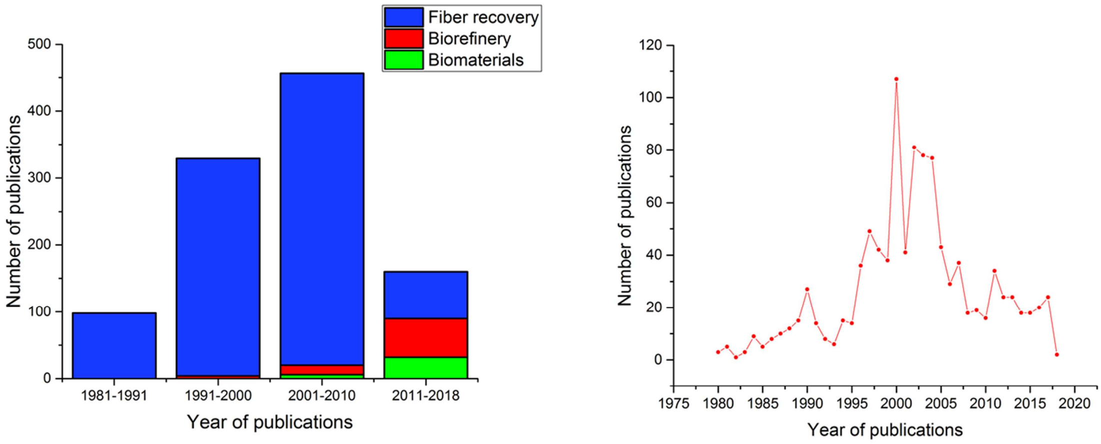

Fig.1 Publications on paper recycling: yearly publications showing increase in 2000 (left), publications by topic or research area (right)

2. Bio-materials synthesised from wastepaper

Paper is primarily a plant/wood based product containing high amount of cellulosic biomass desirable in a diverse range of industries. Cellulose is an abundant natural polymer consisting of crystalline and amorphous regions [4]. Over the past few years, there has been several research breakthroughs in the industrial production of cellulose nanofibers (CNF) and cellulose nanocrystal (CNC) proposing it's use in high added value applications [5, 6]. Wastepaper has been used in the literature for the extraction of CNC/CNF, preparation of polyhydroxyalkanoate (PHA), carboxymethyl cellulose and polymer composite matrix. Meanwhile materials prepared from the synthesis of wastepaper have also been explored for producing high performance electronic components such as supercapacitors.

2.1. Cellulose nanocrystals

Cellulose contains amorphous regions known as hemicellulose which can be removed by acid hydrolysis or mechanical grinding resulting in the formation of a highly crystalline or semi crystalline structure exhibiting elastic modulus as high 137 GPa. Cellulose nanocrystal has been produced from various wastepaper sources such as: old news print (ONP), recycled newsprint (NP), old corrugated container (OCC) and office waste paper (OWP). Cellulose can be obtained from pre-treatment of the wastepaper with sodium hydroxide (NaOH) to remove hemicellulose and bleaching with sodium hypochlorite (NaClO) to remove lignin. Nanocellulose crystals can be prepared with the renowned sulfuric acid hydrolysis method or enzymatic hydrolysis. The resulting properties of the CNCs derived from wastepaper is compiled in Table 1.

Table 1. Characteristics of CNCs prepared from various wastepaper sources

Source Material	**Diameter (nm)**	**Length (nm)**	***CrI*[1] (%)**	**Treatment**	**Ref**
ONP	3 - 10	100 - 300	75.9	Alkali and acid hydrolysis	[7]
NP	5.8±2.2	121±32.5	82.0	Alkali and acid hydrolysis	[8]
OCC	15-80	100-400	57.8	Enzymatic hydrolysis[2]	[9]
ONP	2.9±0.99	371±74	92.6	Direct acid hydrolysis	[10]
NP	3.26±2.90	218±49	93.4	Direct acid hydrolysis	“
NP-B	4.40±3.91	356±137	94.8	Alkali and acid hydrolysis	“
OWP	33±5	238±72	84	Alkali and acid hydrolysis[3]	[11]
OWP	32±5	196±61	73	Alkali and acid hydrolysis[4]	“

The dimension of the wastepaper CNCs obtained above are comparable to CNCs prepared from material sources such as cotton, tunicate, bacteria, ramie and sisal. The CNCs derived from some wastepaper showed aspect ratio between 10-70. CNC vary from 10-100nm in length and 4-70nm in diameter although the crystallinity of CNC (95.5%) derived from pure cellulose is higher. It was shown that with direct acid hydrolysis and no bleaching of the newsprints the CNCs produced had similar properties to bleached newsprint (NP-B) [10]. Thus, Waste paper presents a cheap source for CNC production especially in application whereby the presence of ink, colouring and impurities is of less importance.

2.2. Composite reinforcement

Fibers are commonly used as reinforcing materials in composite applications because of their high aspect ratios which provide high tensile strength and other material properties. Wastepaper (WP) was used as a filler in samples of polyester (P) and polyurethane (PU) as a matrix, with matrix to filler ratio of 20:80 [12]. The tensile strength for the P/WP composite reduced from 17.8±0.5 MPa to

[1] Crystallinity index
[2] 60% phosphoric acid was used for pre-treatment prior to enzymatic hydrolysis and sonication
[3] 2 wt. % sodium hydroxide solution treatment
[4] 7.5 wt. % sodium hydroxide solution treatment

4.4±2.5 MPa whilst modulus increased from 2286±751 MPa to 3144±248 MPa. Nevertheless, another study with 50:50 P/WP ratios reported a significant increase of 30% in the material tensile strength [13] which signifies that there might be an optimum point whereby further addition of wastepaper causes reduction in tensile strength. For the PU/WP, composite strength increased slightly from 1.2±0.2 MPa to 7.8±0.7 MPa whilst elastic modulus increased greatly from 13±3 to 741±113. The mechanical properties in both studies were reduced by 20-30% with increasing moisture uptake due to high cellulose content. The interfacial adhesion of wastepaper in polymer composite matrix can be enhanced by polymer grafting. Maleic anhydride grafted linear low density polyethylene (LLDPE-g-MA) reinforced with WP composites showed significant improvement in tensile strength (88%) and elastic modulus (409%) caused by increased interaction between the wastepaper hydroxyl groups and the anhydride groups [14]. CNC of 20 nm -60 nm widths obtained by acid hydrolysis of wastepaper were used to substitute carbon black for natural rubber (NR) reinforcement which showed few effects on the mechanical properties and improved the processing properties of the NR [15]. Inorganic fillers in recycled paper such as kaolin clay and precipitated calcium carbonate (PCC) have been replaced with CNF which were proven to have a higher bursting and tensile strength [16, 17]. Waste paper fibres have also been incorporated into building materials such as concrete, mortars, bricks and cement based composites [18–20].

2.3. Bio-polymer

CNC films exhibit high transparency, light weight, biodegradability and barrier properties proposing them for packaging applications [21]. Transparent CNC films prepared from OWP were used to coat poly-ethylene terephthalate (PET). The coating with CNC resulted in improved water vapour barrier thought to be beneficial for elongating the shelf life of packaged food products [22]. It was also found that some films appeared dark however still had higher transparency (65%) than others which appeared clear (59%). Sodium alginate/carboxymethyl cellulose (NaCMC) bio-composite films were prepared from old NP. Although the tensile strength of the films (2MPa) was lower than LDPE and HDPE, the materials were deemed suitable for low mechanical packaging applications [23]. The enzymatic hydrolysis of office waste paper produced fermentable sugars used in preparation of Poly (3-hydroxybutyrate) (PHB), a short chain length PHA [24]. Polyhydroxyalkanoates (PHAs) are a family of biodegradable polyesters produced by the microorganisms synthesised in presence of excess carbon source [25]. Although PHAs have their limitations in comparison to synthetic plastics which include high production cost, incompatibility with legacy processing techniques and chances of thermal degradation, improved mechanical properties have been identified when blended with other polymer materials or after chemical modification.

2.4. Electronic components

Hybridisation of Li-ion batteries with electrochemical capacitors require the use of carbonaceous materials with high surface area such as graphene, activated carbon, biomass derived activated carbon for electrode material [26, 27]. Such Li-ion hybrid electrochemical capacitors (Li-HEC) are regarded as potential avenue for efficient energy storage systems. Porous carbon derived from hydrothermal processing and pyrolysis of OWP was used as a cheap source for cathode material in Li-HEC. The material had a high surface area porosity of 2341m^2/g with energy storage capacity of 61 Wh Kg^{-1} [28]. Wastepaper converted into graphene-tethered carbon fibre composite paper (GCCP) demonstrated high electrical conductivity and electrochemical stability for electrodes [29]. Flexible supercapacitors were derived from anchoring reduced graphene oxide-manganese dioxide (RGO-MnO_2) onto OWP. The electrode material exhibited energy storage capacity of 19.6 Wh Kg^{-1} and was proposed for wearable electronic device [30]. Cyanoethyl cellulose derived from waste paper can also be used for organic field-electric transistors (OFETs) [31, 32], such organic based components are receiving attention for their low-cost, ease of processing, flexibility and lightweight circuits suitable in applications for ultra-low power electronics such as radio frequency identification (RFID) tags, biodegradable electronics for medical implant, sensor devices and so on.

3. Wastepaper derived biofuels

The biofuel potential of wastepaper is estimated to replace up to 5% of fossil fuel consumption [33]. In recent studies, the use of wastepaper as feedstock for various energy production such as ethanol, methane and hydrogen has shown promising results.

3.1. Bio-ethanol from wastepaper

Wastepaper has been used as feedstock in anaerobic digestion to produce bio-ethanol and methane through enzymatic hydrolysis [34, 35]. Enzymatic hydrolysis uses enzymes to degrade the cellulose and lignin which produces reducing sugars such as glucose, the fermentation of the glucose produces bio-ethanol which can also be converted to methane [36] as illustrated in Fig. 2.

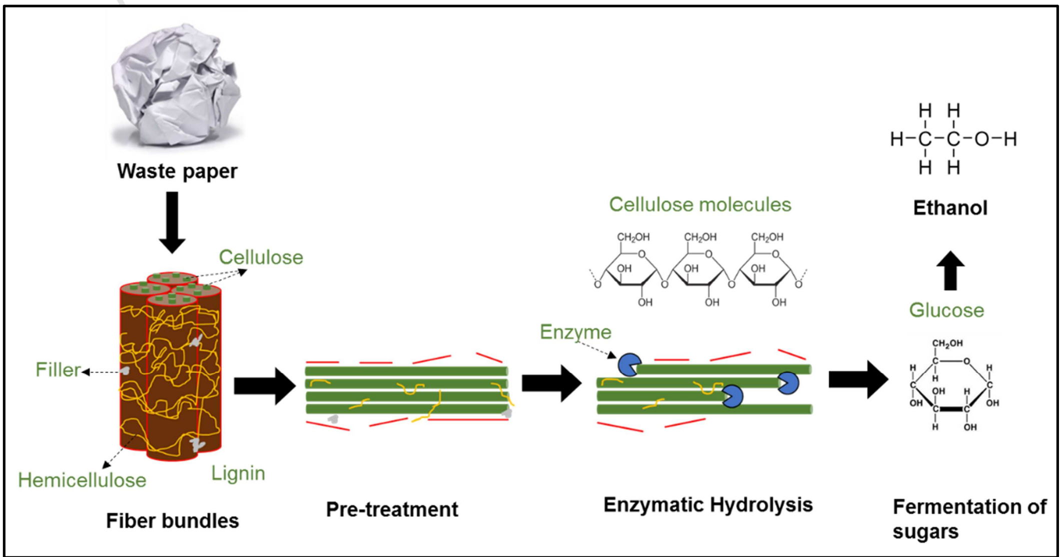

Fig.2. A schematic of the enzymatic hydrolysis of wastepaper to produce fermentable sugars.

The efficiency of hydrolysis process on glucose yield is affected by costly enzymes and long retention times. Recently pre-treatment methods have been shown to improve process yield by disrupting the crystalline structure and increasing the surface area. This enhances the enzymes accessibility to cellulose and reduces enzyme loading. Chemical and physical pre-treatment methods were evaluated to show their effect on glucose release after enzymatic hydrolysis of WP. The maximum glucose yield after milling the WP to 10 mesh was < 0.5 g/L whilst 25 g/L was obtained after sulfuric acid treatment leading to reduction of enzyme loading by 50% [37]. Nevertheless, another study showed that mechanical pre-treatment of WP with Hollander beater prior to enzymatic hydrolysis increased methane yield in anaerobic digestion by 21% at 254 mL/g VS [38], Nishimura *et al* reported higher methane yield of 270.5 mL/g VS when yeast was used for presaccharification prior to simultaneous scarification and fermentation (SSF) [39, 40].

3.2. Hydrogen production

Hydrogen gas is a clean fuel source due to its non-polluting nature during combustion. Biological production of hydrogen through dark fermentation is less energy intensive compared to steam reforming of hydrocarbons and water electrolysis [41]. Fermentation of sugars derived from acid hydrolysis of wastepaper has been proven to produce hydrogen gas. However a limitation in hydrogen yield is the inhibition of the fermentation process caused by furfurals generated during acid hydrolysis stage [42]. Activated carbon has been effective for the removal of Hydroxymethylfurfural (5-HMF) from media after the hydrolysis of wastepaper [43] another study also used lime treatment to reduce 5-HMF from 2.48 g/L to 0.35 g/L [44]. Botta *et al*, applied rumen fluid as an inoculum in wastepaper fermentation, this enriched fermentative spore forming bacterial capable of producing hydrogen. Additionally acid pre-treatment of the rumen fluid reduced the hydrogen consuming cultures [45].

4. Conclusions

Recycling of paper offers several benefits including cost-effectiveness of the process and new emerging market applications. This article highlights that the future of paper recycling will go beyond fiber recovery to recycling wastepaper into functional materials and biorefinery feedstock of higher value. The most prevalent method in the literature for converting wastepaper into CNC is the acid hydrolysis process, however from green chemistry perspectives, more sustainable approaches such as ionic liquid treatment and mechanical refining requires investigation for CNC production from wastepaper. From the energy perspectives, whilst derivation of methane from AD of wastepaper is not especially new, efforts such as pre-treatment, presaccharification and co-digestion have been applied in the literature to improve the process yield and plant economic feasibility. On the bio hydrogen production from wastepaper, the dark fermentation produces mixed gas streams which require improved gas separation technologies to obtain pure hydrogen.

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